Engaging Partners to Evaluate Plastics Loading to the Pamlico Sound from Urban and Rural Lands via the Neuse River in North Carolina

Final Report

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Executive Summary

Recent research has revealed the ubiquity of plastics in our natural environment- microplastics have been found on every continent and in some of the most pristine areas of the planet. However, little sampling and quantification of plastics in North Carolina coastal rivers and estuaries has been conducted. Recent research results vary considerably, both spatially and temporally, and sample collection procedures and reporting methods also differ widely. Seeking to characterize plastic pollution in North Carolina, North Carolina State University (NCSU) and NC Sea Grant applied a variety of field sampling protocols for both macro and micro plastics at fifteen stream gage locations throughout the Neuse River Basin from Raleigh to Craven County from July 2020 to August 2021. This study effort completed the first sampling of microplastics (<5 mm) for North Carolina freshwater rivers and streams. A range of flow conditions (i.e., low flow, storm flow) were captured. Microplastic samples were collected using a 335-micron net following National Oceanic Atmospheric Administration (NOAA) and United States Geological Survey (USGS) methods. The microplastics were extracted from the samples and analyzed using Fourier-transform infrared spectroscopy (FTIR) at the Plastic Ocean Project Lab in Wilmington, NC. Results indicated that microplastic particles were present at all 15 locations. The most common types were polyethylene and polystyrene. The concentrations of microplastics were widely variable across season and flow conditions. The data were analyzed using streamflow, land cover and population variables to determine what factors may influence microplastic concentrations and to determine the relative contribution of plastic loading from different land uses throughout the Neuse River Basin. Microplastic concentrations ranged from 0.02 to 221 particles per cubic meter with the highest concentrations observed in the most developed catchments during stormflow conditions. We estimate that about 670 million microplastic particles larger than 335 µm enter the Pamlico Sound from the Neuse River Basin each year.

Macroplastics were sampled in the upper portion of the river basin using three methods. First, regular collections of trash were completed from a sample grid that included both the channel and the floodplain. These collections were completed at seven streams with a range of development conditions and watershed sizes. Second, trash was captured during stormflow at two highly urbanized streams using a trap composed of a basket and boom system modeled after the copyrighted Litter Gitter[©] device. Third, visual counts of floating trash were completed during stormflow events. The visual observations were made from bridges at two large tributaries and at one small highly urban stream in Raleigh. Floating trash was nearly all plastics and plastics also dominated the litter captured during storm flow using the trash collection boom and basket (96%). Styrofoam pieces were the most common litter type observed using these two sampling methods. Grid samples in contrast contained a more diverse trash profile with plastics comprising about 74% of all samples collected. Urban streams were found to produce much higher counts of trash and macroplastics. Aluminum cans and glass fragments were the most common items among the non-plastic trash. All three macroplastic sampling methods show that the bulk of trash washing into our streams is plastics. In addition, comparisons of trash types and counts by sampling method indicate that only a small fraction of the total trash load is being deposited on the floodplain or channel bed or entangled in debris in the riparian corridor during storm events. Rather, plastics are commonly transported downstream during high flows and are

likely flowing into the mainstem of the Neuse River where it will continue to wash into the Pamlico Sound. Visual counts combined with average flow during the sample recorded were used to estimate a total annual load of floating trash of 120,250 pieces for Marsh Creek. Therefore, programs to prevent this litter from being deposited on the ground and washed into the stormdrain system are critical to plastics from entering stormwater systems in urban areas and being transported to downstream rivers and estuaries of critical social, economic and environmental importance.

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1 Introduction

The staggering proliferation of cheap plastics since the end of World War II has resulted in significant environmental concerns, particularly in coastal ecosystems; today plastics make up an overwhelming majority of all marine debris (Zettler et al., 2013). Marine plastics originate in the terrestrial environment from mismanaged waste, plastic production and other sources such as clothing, cosmetic products, and fishing gear. Today, the world produces about 400 million metric tons of plastic every year, discarding about 300 million metric tons (Geyer et al., 2017). Up to 13 million metric tons end up in the ocean (Jambeck et al., 2015). Rivers are the conduits that supply an overwhelming majority of plastics to the coastal environment (Jambeck et al., 2015).

Over the last decade, research has revealed that while plastics appear to degrade into simple compounds in the aquatic environment; this is not the case. Instead, plastics only breakdown into smaller and smaller components, their total mass not substantially reduced (Yonkos et al., 2014). These tiny particles, referred to as microplastics, are now ubiquitous in the aquatic environment; their occurrence documented in rivers, lakes (Baldwin et al., 2016), estuaries, the open ocean, groundwater and even the deep ocean (Choy et al., 2019). Their distribution wraps around the globe from the Arctic to Antarctic (Simon-Sánchez et al., 2019). Microplastics, operationally defined as particles less than 5 mm in size (Arthur et al., 2009), are made up of fibers, pellets, flakes, foams and films (Mendoza & Balcer, 2019). The very slow degradation rate of plastics means that these particles will persist in the environment for centuries.

There are many unknowns regarding the severity or long term environmental and human health impacts of microplastics, but research has clearly demonstrated microplastics pose an increasing threat to marine life (Thompson, 2015). Because of the various colors and attachment of small marine life (e.g., barnacles), microplastics are often mistaken as a food source by a wide range of marine species and birds. For example, microplastics have been found in sea turtles, fish, whales, sea birds, and other marine organisms (De Sá et al., 2018; De Stephanis et al., 2013; Mascarenhas et al., 2004).

The consumption of larger plastics can cause obstructions in wildlife's digestive tracts and there are also potential toxicity concerns. Because of the surface chemical properties of plastics, toxic substances such as persistent organic pollutants (Rochman et al., 2013) and heavy metals (Munier & Bendell, 2018) readily bind to microplastics. Leaching of plastic additives is also a concern (Koelmans et al., 2014). Wildlife can be exposed to these pollutants when they consume microplastics and there is also the risk of negative impacts higher up in the food web due to bioaccumulation of toxins (Teuten et al., 2009).

Research indicates a wide range of microplastic distribution spatially and temporally and sample collection procedures and reporting methods themselves are varied. For example, results have been reported in terms of volumetric density (g/m³), areal density (g/km²) and total particles (number of particles). Globally, reported concentrations have varied from 3.5 particles per m³ in the Ebro River in Spain (Simon-Sánchez et al., 2019) to 108 particles per m³ in the Seine River in France (Dris et al., 2015) and almost 8000 particles per m³ in rivers in China (Lin et al., 2018). In tributaries to the Great Lakes, microplastic concentrations varied from 2 to 32 particles /m³

(Baldwin et al., 2016), but average surface concentrations in the Great Lakes was 43,000 particles per km². Closer to North Carolina, 100 to 1,500 particles per m² were reported for estuaries in South Carolina (Gray et al., 2018) and a wide range of microplastic concentrations (1 to > 560 g/km²) were reported in Chesapeake Bay tributaries. Although not directly comparable to other studies, the Chesapeake Bay results showed higher levels in more populated watersheds, and potential correlation with river discharge. Similar findings were reported elsewhere (Baldwin et al., 2016; Moore et al., 2011).

In order to understand microplastic inputs to coastal waters, larger plastics (macroplastics) should also be quantified as they represent the majority of plastic input to our rivers and break into microplastics. The majority of plastic transport occurs during high flow events. Research from the Seine River in France showed a tenfold increase in plastics transport during high discharge events and the number of plastics increased by a factor of 5 from upstream to downstream of Paris. They reported results in the range of 100 to1000 plastic pieces per hour, potentially representing tens of thousands of microplastic particles in the future.

While these previous studies point to a pervasive issue that is drawing increasing attention, the severity of the problem in North Carolina has not been documented. To date, there have not been any studies to quantify the concentration of plastics in coastal waters or estimate the plastic load from riverine systems. Due to the scale of the problem and the continued production of plastic, macro and microplastics pollution will not be solved by cleanup efforts alone (Kershaw, 2016). Instead, work needs to be focused on reducing the use of plastics and the generation of plastic waste (Abreu & Pedrotti, 2019). These are primarily education, behavior and policy dilemmas.

The primary goal of this study was to take the first step in characterizing and quantifying the annual loading of plastic pollution to our coastal waters from inland sources by examining contributions through the Neuse River watershed to the Pamlico Sound. The secondary goal was to use research results to raise awareness of plastic pollution since quantifying the scale of the problem in a local context has been shown to increase stakeholder engagement and interest (Vorkinn & Riese, 2001). Specific objectives of this project included:

- 1. Determine microplastic concentrations in streams throughout the Neuse River Basin.
- 2. Estimate the plastic microplastic loading rate from the Neuse River to the Pamlico Sound
- 3. Characterize and evaluate the relative source of microplastics loading based on land use (urban vs. rural).
- 4. Estimate macroplastic loading to streams and evaluate the relative loading from urban and non-urban areas in the upper and middle Neuse River Basin.
- 5. Test and refine methods and strategies to sample for macro and micro plastics in North Carolina coastal draining rivers to estimate loading rates.

2 Methods

2.1 Study Sites

Field sampling for microplastics was conducted at 15 locations distributed throughout the Neuse River Basin. Grid sampling of macroplastics and other trash was conducted at 7 of the 15 sites that were located in the upper portion of the watershed. Visual sampling of floating trash including macroplastics was conducted at three of the study sites. A map indicating the location of each study site is shown in Figure 1. The type of sampling that was conducted at each site is provided in Table 1 below.

The Neuse Basin drains an area of almost 5,600 square miles that flows into the Pamlico Sound, North Carolina's most important fishery. The Basin is home to over 2.5 million people, mostly concentrated in the highly developed upper watershed, while the lower watershed is dominated by agricultural and forested land uses. Sample locations were strategically selected to include a range of drainage area sizes, a variety of land use (i.e., urban and rural), and to encompass locations throughout the basin. All the sample locations were near US Geological Survey (USGS) streamflow gages. All sampling was conducted between August 2020 and July 2021.

| Site | Sample Location | % | Drainage Area | Macroplastic |
|--------|------------------------------|-----------|--------------------|--------------------|
| Number | | Developed | (km ²) | Sampling Method |
| 1 | Rocky Branch in Raleigh | 93 | 3.0 | Trap, Grid |
| 2 | Marsh Creek in Raleigh | 95 | 18 | Trap, Grid, Visual |
| 3 | Crabtree Creek in Raleigh | 65 | 311 | Grid, Visual |
| 4 | Walnut Creek in east Raleigh | 82 | 78 | Grid, Visual |
| 5 | Swift Creek near Apex | 80 | 54 | Grid |
| 6 | Swift Creek near Clayton | 32 | 298 | - |
| 7 | Middle Creek Near Clayton | 35 | 220 | - |
| 8 | Little River near Zebulon | 10 | 142 | Grid |
| 9 | Neuse River at Clayton | 21 | 297 | - |
| 10 | Little River near Princeton | 10 | 596 | - |
| 11 | Neuse River at Goldsboro | 12 | 6,220 | - |
| 12 | Nahunta Swamp | 7 | 218 | Grid |
| 13 | Contentnea Creek at | 10 | 1,890 | - |
| | Hookerton | | | |
| 14 | Neuse River at Fort Barnwell | 10 | 10,100 | - |
| 15 | Trent River | 4 | 440 | - |

Table 1. Microplastic and macroplastic sampling locations and methods



Figure 1. Sample locations.

2.2 Sampling Methods

2.2.1 *Microplastics*

Trawl samples were collected six times at all 15 sites capturing a range of streamflow conditions at each location. The methods described by Baldwin et al. (2016) were closely followed. A 335 μ m mesh neuston net with a 100 cm wide by 40 cm height frame openings was used to sample the upper 20-25 cm of the streamflow (see Figure 2). Samples were collected while positioned on bridges, boat docks, boats, and by wading instream, depending on the size of the river, access, flow and flood stage at the time of sampling. Trawl times ranged from 3 minutes to 15 minutes depending on the streamflow and clogging of the net. The average velocity of the streamflow entering the net was measured using a velocity meter. The total volume of water filtered through the net ranged from 9 to 134 m³. In addition, two subsurface samples were collected using a smaller version of the neuston net (30 cm diameter).



Figure 2. Sample trawl net with floats and velocity meter attached (a), sampling from a bridge (b), and sampling using cable suspended across the stream channel (c).

2.2.2 Macroplastics

Macroplastics were sampled using the three methods described below.

2.2.2.1 Grid Sampling

At seven study sites, a stretch of stream where litter was visibly accumulating in the channel and floodplain was identified. A rectangular area encompassing the stream and floodplain was marked using survey stakes and a GPS (See Figure 3). Each gridded area was pre-cleaned to establish a baseline. Each site was subsequently cleaned between 2-5 times to remove all newly accumulated trash. All samples were tallied by type (i.e., bottles, bags, etc.), dried, and weighed. Forty-two categories of trash type were identified based on Big Sweep and other trash cleanup collection programs. A field data form was used to record each litter item as it was collected (see Appendices). A Google Form was also developed to upload the litter data into a spreadsheet. A library of weights for plastic bottles, aluminum cans, glass bottles and athletic balls was developed based on size and type to ease the burden of washing, drying and weighing of all litter items. The library of weights for plastic bottles is included in the Appendices.

With the exception of Nahunta Swamp, all litter was collected by project investigators rather than volunteers due to COVID-19 related restrictions. The record of flow that occurred between each grid collection was downloaded from each USGS gauging station. The number of flows that reached or exceeded the estimated bankfull discharge for the watershed area as determined by the North Carolina Piedmont hydraulic geometry regional curves (Doll et al., 2002) were recorded between trash collection dates. In a stable stream, bankfull flow is the maximum discharge that the channel can convey without water overflowing onto the floodplain. Bankfull was selected as the threshold flow that is likely to generate significant transport of debris as this flow will also move significant loads of sediment. A large portion of the garbage was bagged and transported to the NC State University campus where it was washed, dried and weighed. Not all items were transported and weighed due to size, bulk and/or sanitation issues.



Figure 3. Macroplastic sampling grid approach.

2.2.2.2 Trap Sampling

With permission from Osprey Initiative (https://osprey.world/), two prototypes of their patented Litter Gitter© trash collection system were constructed. Each device included a floating basket with a boom system to direct trash into the basket. The boom was assembled from nylon rope

and lobster buoys; the basket was constructed using PVC pipe, PVC coated landscape wire and metal fasteners (Figure 4). The traps were placed in Rocky Branch and Marsh Creek and secured to trees or a landscape stake installed along the streambank. A segment of channel with more laminar flow (i.e. smooth rather than turbulent) was selected to maximize trash capture. The traps remained in the creek for a few months in order to sample several storm events. After each storm event, all trash was removed, cataloged; and later it was washed, dried and weighed. The corresponding peak discharge for each storm event was obtained from the USGS station gaging record.



Figure 4. Removal of trash from the Litter Gitter[©] prototype following a storm event at Marsh Creek

2.2.2.3 Visual Assessment

Visual assessments were conducted at two larger urban tributaries (Crabtree Creek and Walnut Creek) that drain most of the City of Raleigh and one small highly urban creek (Marsh Creek) (Figure 5). Each visual count was conducted for approximately 30 minutes during storm flow events using the Floating Litter Monitoring Application from the European Commission's Joint Research Centre following the methodology outlined by (González-Fernández & Hanke, 2017) and (Van Emmerik et al., 2018). At Crabtree Creek and Marsh Creek, the observer was positioned at a bridge crossing close to our grid sampling site, and the associated USGS gauging station. For Walnut Creek, the visual assessment was conducted from a bridge that was on the opposite side of town from the grid site because of accessibility limitations near the gauge. The observer recorded each piece of litter by type (e.g., styrofoam, plastic bottle, etc.) and approximate size as it was observed floating past. Each entry was georeferenced and time-stamped by the app.



Figure 5. Visual assessment sites in Raleigh.

2.3 Microplastic Laboratory Analyses

Organic material was removed from each sample through chemical digestion using catalyst wet peroxide oxidation and heating to 60° C. After digestion, the sample was rinsed with deionized water through a 100 µm nylon filter. Potential plastic particles and fibers were then visually identified and removed. The remains of the sample were subjected to density separation using a salt solution. Again, suspected plastic particles were individually removed and the surface liquid and buoyant particles were poured off and sieved. The sieved materials were visually inspected again and all suspected plastics were removed and placed into the acrylic container (see examples shown in Figure 6). The remainder of the sample, containing the settled materials, was inspected in the same manner.

Suspected plastic particles were cleaned and then examined using a Wolfe SMZ-161 stereomicroscope. The particles were also categorized as fibers or fragments and analyzed using a Micro Fourier Transform Infrared Spectrometer (Micro FTIR). High resolution scans were collected and the possible identities were compared to multiple polymer and biological libraries within the Omnic Picta[™] software to determine the best fit. For three samples in which more than 2000 potential plastic particles were identified, about 10% of the particles were subjected to FTIR analysis.



Figure 6. Examples of potential plastic particles removed from the sample following chemical digestion and density separation.

2.4 Microplastic Data Analysis

Microplastic concentrations were calculated for each water sample and compared to stream discharge at the time of sampling using the non-parametric Spearman Correlation test. The types of plastics identified were compared between samples and cumulatively for each site. Plastic particles were grouped by polymer type (e.g., polyethylene, polypropylene, etc.) when they accounted for at least 2% of the particles in the sample. Polymers that made up less than 2% were grouped together into a "miscellaneous plastics" designation. The composition of polymer types was calculated for each site and principal component analysis was completed to evaluate commonalities in grouping (i.e., site, season, and streamflow). We examined the influence of land cover, development, and population on median microplastic concentration across the nine tributary watersheds. For development and forest cover data, we relied on the 2019 National Land Cover Database (MRLC, 2022) while population density was obtained from the 2020 United States Census Bureau Data (2022). The Spearman correlation test was used to test for significant relationships.

2.5 Microplastic Loading Estimate

A rough estimate of the total annual microplastic loading from the Neuse River basin to the Pamlico Sound was calculated. Following an approach proposed by Eo et al. (2019), we separated the channel into a surface (top 0.2 m) and subsurface component. Because only two subsurface samples were collected, we assumed the subsurface microplastic concentration was 1/3 of the surface concentration based on the data reported by Eo et al. (2019). The mean microplastic concentration (p/m³) for each site was then multiplied by the total flow volume at Fort Barnwell for 2020 and 2021, and averaged to arrive at a total annual load in particles per year.

2.6 Floating Trash Load Estimates

Each visual observation at the three creeks was associated with the discharge at corresponding stream gage. The number of plastic pieces observed was compared to the streamflow to evaluate if there was a relationship between the number of pieces transported and the flow rate. The data was log transformed and fitted with a power function. The resulting regression equations were used to estimate annual plastic loads for Marsh and Crabtree creeks. To estimate the annual load,

the 15-minute gauge data were downloaded for the period of July 1, 2020 to June 30, 2021. The discharge-loading regression relationships were then used to estimate the amount of trash transported over each fifteen-minute increment and then summed for all storm events. The total-storm-transport was calculated over the time period beginning at the estimated discharge needed to transport litter, to the storm peak discharge. The falling limb of the storm (when flow volume declines) was not included in the trash load estimate as we assumed that the bulk of the trash is flushed out of the watershed by the time of the peak discharge (i.e., the time of concentration for the catchment). The time of the sampling relative to the storm hydrograph (i.e., rising limb, peak or falling limb) was noted to identify potential changes in trash loading that may occur as accumulated litter is washed off the urban landscape and transported through the stream network.

2.7 Comparison of the Trash Profile by Sampling Methods

The three macroplastic sampling methods were compared using data from Marsh Creek, where all three practices were employed – grid, trap, and visual. The total count and the percentage of trash types were normalized based on the sampling effort for each method. Specifically, the grid count was divided by the number of flow events that exceeded bankfull discharge between the sampling events (i.e., the approximate number of times substantial trash volumes were likely transported prior to cleaning). Similarly, the sum of the trap collected trash was divided by the number of storms that were sampled. For the visual count, the total load of trash for each of the six storms sampled was estimated using the methods described in the previous section. This total storm count was then divided by the total count of trash observed during the six-~30 minute sampling periods to calculate a "total storm multiplier". To calculate the total storm count for each trash type, the count for each trash type was divided by the number of visual observations (6) and then multiplied by the "total storm multiplier".

3 Results and Discussion

3.1 Microplastics

Microplastics were present in all of the water samples. Concentrations for the 90 samples collected using the 335 μ m mesh ranged from 0.02 p/m³ to 221 p/m³ with a median of 0.44 p/m³; however, only 5 of the samples had concentrations greater than 10 p/m³ (Figure 7). The highest concentrations were generally observed in the urban areas of the upper watershed (i.e., in and around Raleigh). One sample collected from the Trent River, collected during a dry period with very low flow and stagnant conditions, had a concentration of 11 p/m³. Other samples from this site were collected during higher streamflow and had much lower concentrations, potentially indicating a relationship between drought conditions and localized microplastic accumulation. The concentrations reported here were generally comparable to or lower than previous studies using analogous methods in areas with similar waste management infrastructure. Dris et al. (2015) reported a range of 0.28 to 0.47 p/m³ in the Seine River in Paris. Similarly, Lechner et al. (2014) reported a mean concentration in the Danube River in Germany of 0.32 p/m³, with a maximum of 142 p/m³. In the United States, Baldwin reported slightly higher average levels with a median of 1.9 and mean of 4.2 p/m³. in tributaries to the Great Lakes. In rivers of heavily urbanized Milwaukee WI, Lenaker et al. (2019) reported a range of 0.5 to 11.6 p/m³.



Figure 7. Water sample results (335 µm) across the Neuse River Basin.

3.1.1 Impact of Streamflow

Six trawl samples (335 µm mesh) were collected across a range of flows at each location. The relationship between stream discharge and microplastic concentration was not consistent across all sites (Figure 8). For the smaller, urban streams in the upper basin (Rocky Branch, Marsh, Walnut, Crabtree and Swift (Apex) Creeks), there appears to be a trend of increasing microplastic concentration with increased discharge; however, this correlation was only statistically significant for Rocky Branch and Marsh Creek (the most developed catchments). There was no clear relationship with flow for the other smaller catchments or along the mainstem river segments. A larger sample size could improve our understanding of the relationship between flow and microplastic transport.

The relationship between discharge and microplastic concentration for urban streams potentially indicates that there is substantial microplastic transport occurring during storm events. The five samples with the highest microplastic concentrations were collected at or near the peak of the hydrograph following intense rainfall events in three urban creeks (Rocky Branch, Marsh Creek, and Walnut Creeks). These sample concentrations were an order of magnitude or higher than the samples collected at baseflow. Baldwin et al. (2016) also reported the highest concentrations of microplastics in samples collected from urban sites during high streamflow, and Gündoğdu et al. (2018) reported increased concentrations in estuaries following flooding events. There are a few factors that likely contribute to this increased microplastic transport during stormflow. First, similar to other types of pollution, microplastics that have built up on impervious surfaces may be flushed into streams during initial runoff from a rain event (Werbowski et al., 2021). Second, microplastics in stream sediments can be mobilized during high flow events (Hurley et al., 2018).



Figure 8. Trawl (335 µm mesh net) collected microplastic concentration versus stream discharge. The Spearman correlation coefficient and associated P-value are shown for each location.

3.1.2 Watershed Attributes

There was a strong positive correlation (r > |0.6|) between median microplastic (>335 µm) concentration and development and population density, and a negative correlation with forest cover, for the tributary catchments (Figure 9). The correlations were only significant for forest cover. This intuitively makes sense given that more plastics are consumed and more waste is generated in more populated areas. However, while some previous studies have reported a correlation between microplastic concentration and population density (Baldwin et al., 2016; Yonkos et al., 2014), others found no such relationship (Dikareva and Simon, 2019). Since our study covers only a few sites (9 catchments) and a relatively small number of samples (6 per site), it does not resolve the current disparity.



Figure 9. Relationship between median microplastic (>335 μ m) concentration and catchment developed land (a), population density (b), and forest cover (c) for the tributary catchments. The spearman correlation coefficient and associated p-value are included for each plot.

3.1.3 Polymer Identification

Of the 7557 particles analyzed using FTIR, 6303 (83%) were confirmed to be synthetic plastics, resins and paints (e.g., urea formaldehyde, polyurethane), plastic additives, or bioplastics (e.g., Rayon, Cellophane, Cellulose Acetate). The majority of the plastic particles found were fragments, films and foams; only 20% were fibers. Over 150 unique synthetic compounds were identified through FTIR analysis, confirming that microplastics make up a diverse and complex mixture of different chemical compounds, not just the most common polymers (Rochman et al., 2019).

Polyethylene (PE) was the most common polymer type (~30% of particles), and was found in 98% of the samples. PE is also the most produced plastic in the world (Jeremic, 2014). Polyethylene is widely used for plastic bags, films, packaging, and bottles. Polypropylene was the next most plentiful polymer (18%) and was present in 70% of the samples. Polypropylene is often used for food containers and other packaging. Synthetic fibers are used for carpeting, and rope. The third most common polymer was polystyrene (15%) and was found in 77% of the samples. Polystyrene is widely used for packaging, insulation, and foam food/drink containers. The remainder were made up of polyvinyl polymers, bioplastics, rubber and miscellaneous polymers (acrylonitrile butadiene styrene, acrylics, ethylene-vinyl acetate, polyamides, rubber (~1% of particles) (Table 2). Various thermosetting plastic resins were also found including alkyd resins, epoxy resins, phenoxy resins. The most common thermosetting resin identified was urea formaldehyde (~6%), which is the adhesive often used to manufacture oriented strand board (chipboard), particleboard, and plywood (Hiziroglu, 2018). These particles were often identified as urea formaldehyde in combination with chipboard or melamine. Paints and other sealants were also present, including polyurethane and latex paints.

Polyethylene terephthalate (PET) was present in about 6.5% of the samples. PET is used in single use plastics and is the most common synthetic fabric (i.e. polyester). About 5.5% of the particles were identified as copolymers of other common plastic polymers, including poly(ethylene: vinyl chloride), poly(ethylene: propylene), polyethylene/ethyl acrylate. These common polymers are often combined to create new compounds with desirable properties inherited from its constituents.

Some of the particle's chemical identity based on FTIR analysis did not match their physical appearance under the microscope. This was likely because the spectral signature of plastics changes as the plastic surface ages (Castelvetro et al., 2021). For example, some particles in our samples were most closely matched to and therefore identified as food additives (e.g., Sorbitan monopalmitate, Sorbitan monostearate). The FTIR signature of these particles was also very similar to the signature of degraded polyethylene reported by Castelvetro et al. (2021). Under our microscope these particles resembled a plastic film, we therefore assumed the particles were either degraded polyethylene, or plastic film coated with a food additive. Other particles, identified as cork using FTIR, also appeared to be films when examined under the microscope. We assumed these particles were polyethylene based on the similar spectral signature and our visual inspection.

Some of the rarer particles included several thermoplastics used for nonstick coatings (polytetrafluoroethylene and Xylan). Particles in one sample collected from Crabtree Creek in Raleigh were identified as Bakelite. This is the first plastic ever made from synthetic components, was patented in 1909, and mostly phased out after World War II. Interestingly, this site was undergoing sewer line construction work that involved excavation of the streambed, which may have resulted in suspension of legacy plastic particles.

| Polymer Type | Overall Proportion | Fraction of Samples w/ Polymer Present |
|--|-----------------------|---|
| Polyethylene (HDPE, LDPE, LLDPE) | 30.3% | 98% |
| Polypropylenes | 18.0% | 70% |
| Polystyrenes | 15.0% | 77% |
| Resins and plastic additives (e.g., Polyurethane, Paints, Plastic additives, pigments) | 7.4% | 73% |
| Polyethylene terephthalate (PET) | 6.4% | 42% |
| Urea Formaldehyde Resin (used as adhesive in particle board, chipboard and plywood) | 6.3% | 69% |
| Misc. Plastics (ABS, PA, PC, PMMA, acrylics, rubber) | 5.9% | 61% |
| Various Copolymers (e.g., poly(ethylene: vinyl chloride), poly(ethylene: propylene)) | 5.6% | 62% |
| Polyvinyl polymers (e.g., PVA, PVC) | 3.1% | 32% |
| Bioplastics (e.g., Cellophane, Rayon, Cellulose Acetate) | 2.0% | 37% |

Table 2. Proportion and presence of polymer types in trawl (335 µm mesh) water samples.

3.1.4 Polymer Distribution

Overall, there was substantial variability in both the types of plastics collected at each site and the composition of plastics collected from sampling event to sampling event (Figure 10 and Figure 11). Principal component analysis (PCA) indicated that streamflow, season, and location

were not strong contributors to the variability in types and composition of plastics (Figure 12), although streamflow appeared to relate to the greatest variability in plastic composition. The lack of consistency in the composition of microplastics within a given site likely indicates complex factors that contribute to highly variable sources of microplastics, both spatially and temporally.



Figure 10. Cumulative distribution of polymer types by sample location.



Figure 11. Relative distribution of polymer types by sample location and date.



Figure 12. Principal component analysis of polymer composition grouped by a) streamflow, b) sample date, and c) site.

3.1.5 Microplastic Loading

We estimate that about 670 million microplastic particles greater than 335 µm enter the Pamlico Sound from the Neuse River Basin each year. This is a very coarse estimate of microplastic loading with a high degree of uncertainty. Reducing the uncertainty would require a more extensive sampling study to improve the understanding of spatial and depth variability of microplastic dynamics in rivers. Recent studies have revealed some of the variability and uncertainty. For example, Lenaker et al. (2019) reported widely variable results and did not see a consistent relationship between microplastic concentration and depth, and Dris et al. (2018) reported variability both with depth and across the width of the stream channel.

3.2 Macroplastics

A total of 31 grid samples were collected at the seven tributary streams selected for macroplastic sampling; 32 Litter Gitter© samples were collected at the two small very urban creeks (Rocky Branch and Marsh Creek); and a total of 16 visual observations were completed at three of the urban study creeks. The total samples collected and the total pieces of plastic litter counted at each stream are provided below in Table 3.

| Sample Location | % Developed | Grid Method | # Pieces | Trap Method | # Pieces | Visual Method | #Pieces |
|-----------------------|----------------|----------------|-------------|----------------|-------------|------------------|---------|
| Marsh Creek | 95 | 5 | 677 | 15 | 6295 | 6 | 655 |
| Rocky Branch | 93 | 5 | 490 | 17 | 1019 | | |
| Walnut Creek | 82 | 5 | 442 | | | 1 | 248 |
| Swift Creek near Apex | 80 | 4 | 340 | | | | |
| Crabtree Creek | 65 | 5 | 60 | | | 9 | 420 |
| Little River | 10 | 2 | 34 | | | | |
| Nahunta Swamp | 7 | 5 | 4 | | | | |
| Total | | 31 | 2047 | 32 | 7314 | 16 | 1323 |

Table 3: Sample counts and total pieces of plastic litter (including rubber) for each of three sampling methods for each study location.

3.2.1 Grid Sampling

3.2.1.1 Pre-cleaning

All seven grids were pre-cleaned in July and August of 2020 to remove all legacy trash. 1,565 pieces of trash were collected at the seven study sites. Sixty-five percent of this trash (1020 pieces) were plastics. In addition, several large items were collected from the floodplain of Marsh Creek including six tires, an automotive bench seat, several car bumpers, two piles of flexible plastic sheeting and many fluorescent glass tube light bulbs. This large debris indicated that dumping of trash has likely occurred in or very near the floodplain at this location. In general the trash from the pre-cleaning at all sites was predominantly composed of styrofoam pieces, plastic bottles, aluminum cans, plastic film, glass fragments, food wrappers, plastic bags, toys, balls and hard plastic pieces. These items comprised about 78% of the pre-collected trash from the study sites. A distribution of the litter by number of pieces is provided in Figure 13 below. Nearly half of the pre-cleaning trash (46%) was collected from Marsh Creek. Much less garbage was collected from the two less developed streams (Nahunta Swamp and Little River) as compared to the other five more developed streams. Table 4 shows the amount of trash and the percentage of total trash collected form the seven study sites.



Figure 13. Distribution of trash collected during pre-cleaning of seven study sites.

Table 4. Distribution of trash collected by stream during grid pre-cleaning. The percent of development in the watershed is also included.

| Creek | Trash Count | % of Total | % Developed |
|---------------------|--------------------|------------|-------------|
| Marsh Creek | 713 | 45.6% | 95 |
| Crabtree Creek | 385 | 24.6% | 65 |
| Rocky Branch | 161 | 10.3% | 93 |
| Walnut Creek | 159 | 10.2% | 82 |
| Swift Creek | 118 | 7.5% | 80 |
| Nahunta Swamp | 16 | 1.0% | 7 |
| Little River | 13 | 0.8% | 10 |
| Total | 1565 | | |

3.2.1.2 Grid Samples

A total of 2,676 pieces of trash were collected during the project (31 grid samples). The bulk of this trash had a cumulative dry weight of approximately 92 pounds. This total weight does not include bulky and heavily soiled items. Most of the trash was collected from 4 of the 7 study sites. Two of the three sites with the least trash have very low levels of development within the watershed (Littler River at 10% and Nahunta Swamp at 7%). Relatively little trash was also collected at Swift Creek near Apex despite a high level of watershed development (80%). However, there is a large lake upstream of the Swift Creek sampling location which may capture

trash and prevent downstream movement. Over 76% (2,047 pieces) of this litter was classified as plastics (including rubber). Twenty-nine different categories of plastic trash were encountered. The bulk of the plastics collected included styrofoam pieces, food wrappers & labels, beverage bottles, bags, hard plastic pieces and food containers. The distribution of plastic litter items collected are shown in Figure 14. The top 12 items (Figure 15) comprise 94% of all the plastic trash collected in the grid samples.



Figure 14. Distribution of plastic items collected during grid sampling (n=33) at seven study sites.

Seventy-two percent of the non-plastic trash collected in the grid samples was aluminum cans and glass fragments. Figure 15 below provides the item counts for each of the 12 categories of non-plastic trash collected.



Figure 15. Cumulative piece counts for 12 non-plastic trash items collected from grid samples (n=32) at the seven study sites.

Figure 16 shows the timing of the pre-cleaning and subsequent cleanups of the grid areas relative to the record of discharge downloaded from each USGS gage station. The number of storms exceeding bankfull discharge prior to each gird cleanup are summarized in Table 5. The trash collected per grid in acres divided by the number of cleanups for each site is also provided in Table 5. The streams are sorted from those with the highest percent watershed development to the lowest. It is clear that development has a strong influence on the loading at these seven streams. It also appears that the watershed size and the number of significant storms that occurred prior to each cleanup impacted the amount of trash that was collected.



Figure 16. USGS flow gage results for seven study sites. The timing of the pre-cleaning of the grid area (red bar) and the subsequent grid sampling events (blue bars) are also indicated.

| Table 5. | Summarv o | f Trash | Counts f | for | Trash and | l Macro | plastics | collected | from | grid san | ınlin | g |
|----------|---|---------|----------|-----|-----------|---------|----------|-----------|------|----------|---------------------|------------|
| | ~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~ | / | J | | | | r | | | | · · · · · · · · · · | o ' |

| Sampling Site | Total Macroplastics | Total Non-Plastics | Total Trash | # Clean-ups | # Storms ≥ Bankfull Q | Grid Area (acres) | % Developed | Watershed Area | Trash /acre / # Cleanups |
|----------------|---------------------|--------------------|-------------|-------------|--------------------------|-------------------|-------------|----------------|-----------------------------|
| Marsh Creek | 677 | 69 | 746 | 5 | 18 | 0.53 | 95 | 6.9 | 280.5 |
| Rocky Branch | 490 | 94 | 584 | 5 | 14 | 0.35 | 93 | 1.2 | 337.3 |
| Walnut Creek | 442 | 25 | 467 | 5 | 4 | 0.75 | 82 | 30.1 | 125.0 |
| Swift Creek | 34 | 4 | 38 | 4 | 6 | 1.68 | 80 | 20.8 | 5.7 |
| Crabtree Creek | 340 | 400 | 740 | 5 | 2 | 1.61 | 65 | 120.1 | 92.2 |
| Little River | 4 | 1 | 5 | 2 | 1 | 1.36 | 10 | 54.8 | 1.8 |
| Nahunta Swamp | 60 | 36 | 96 | 5 | 4 | 0.99 | 7 | 84.2 | 19.3 |
| Total | 2047 | 629 | 2676 | | | | | | |

3.2.2 Trap

A total of 7,449 pieces of trash were captured from 32 storms using the trap at Rocky Branch and Marsh Creek. Most of this collected trash was dried and then weighed reporting a total of approximately 101.2 pounds. Bulky, heavily soiled or unsanitary items were not dried and weighed. 7,314 pieces (98%) were plastics (including rubber) and represented 27 categories of plastics. Over half (55%) of the items were styrofoam pieces. Food wrappers, beverage bottles, bags, hard plastic fragments and cups and food containers were also common. The distribution of the plastic trash collected is provided in Figure 17 below. The top 12 items, which are labeled in Figure 17, comprise 98.7% of all the plastics.



Figure 17. Distribution of plastic items collected by trap (n=32) at Marsh Creek and Rocky Branch.

Only 135 non-plastic items including aluminum cans, glass bottles, paper, and tin cans were captured. The piece count for the 12 non-plastic items is shown in Figure 18 below.



Figure 18. Piece counts for 12 non-plastic trash items collected from litter gitter samples (n=32) at Rocky Branch and Marsh Creek.

The trash count versus peak discharge for the associated storm event for each trap collection are graphically compared for both Rocky Branch and Marsh Creek in Figure 19 below. The Rocky Branch data indicate less trash collected during larger flow events. This may be due to large logs and debris blocking the trash collection basket and/or more turbulent flow typical of higher flow velocities that resulted in trash overtopping the boom and thereby bypassing the basket. It appears that our Litter Gitter[©] prototypes are more effective at intercepting trash and preventing its downstream movement during low to moderate storm events. This indicates the importance of preventing trash from entering the stormwater system because once in the waterway it is difficult to capture and prevent the further downstream transport of plastics.



Figure 19. Peak Discharge versus Trash Count for litter gitter collections at Rocky Branch and Marsh Creek.

3.2.3 Visual Counts

Visual counts of trash were collected from three creeks including Crabtree, Marsh and Walnut. 425 pieces of trash were counted during 9 approximately thirty-minute observations at Crabtree Creek. The observations occurred during a range of flow conditions (Q=281 to 2,404 cfs). Two counts occurred at different times on the same storm (9/25/2020) and at two separate locations along the creek approximately 3 miles apart (Anderson Dr. and Crabtree Valley Mall). Most of

the observations occurred during the rising limb or near the peak of discharge for each storm event (see figures 20-22). All but 5 pieces observed at Crabtree Creek were plastics.



Figure 20. USGS flow for each storm event when visual observations were made (red vertical lines) for Crabtree Creek at Anderson Drive and at Glenwood Ave on 9/25/2020.



Figure 21. USGS flow for each storm event when visual observations were made (red vertical lines) for Marsh Creek at Stoneybrook Dr.



Figure 22. USGS flow for each storm event when visual observations were made (red vertical lines) for Walnut Creek at Rose Lane.

Styrofoam pieces, other plastic pieces, and plastic bottles were the most dominant types of floating trash that were observed during storm flow. These three items comprise 82% of all the trash observed at Crabtree, 79% of the trash at Marsh Creek, and 81% of the trash at Walnut Creek. Figures 23-25 below provides a breakdown of the trash that was counted at Crabtree, Marsh and Walnut creeks.



Figure 23. Trash composition for 9 visual counts made during 8 storm events on Crabtree Creek.



Figure 24. Trash composition for 6 visual counts made on Marsh Creek.



Figure 25. Trash composition for 3 visual counts made on Walnut Creek

3.2.3.1 Plastic Loading Estimates

The number of pieces of floating trash transported per minute compared to the average flow recorded at each USGS gage during the visual observations for Crabtree, Marsh and Walnut Creeks is shown in Figure 26. The Crabtree Creek data produced the strongest positive trend between discharge and the quantity of trash observed. ($r^2 = 0.91$), The Marsh Creek data also provided a fairly strong positive trend ($r^2 = 0.74$). A trend was not evaluated for Walnut Creek since only three observations were made at that location. The relationships for Crabtree and Marsh creeks were used to estimate the total annual floating trash load for both creeks. Based on the observations for Crabtree Creek, minimal amounts of trash start to be transported at around 50 cubic feet per second (cfs) with more significant transport of trash occurring when discharge begins to reach 150 to 180 cfs. Therefore, only flows above 150 cfs on the rising limb of the storm hydrograph were used to estimate the total annual trash load for Crabtree Creek. At Marsh Creek trash transport appears to initiate at around 20 cfs, so flows above 20 cfs on the rising limb of the storm hydrograph were used to calculate the total annual load of floating trash. For Crabtree Creek the total estimated annual load of floating litter was 47,870 pieces; and for Marsh Creek it was 120,250 pieces. It should be noted that Marsh Creek flows into Crabtree Creek at a point below the sampling location on Crabtree Creek. In addition, there is a large lake upstream on Crabtree Creek (Lake Crabtree), which may capture macroplastics and reduce downstream loading.



Figure 26. Number of pieces of trash observed versus USGS recorded discharge for Crabtree Creek (n=9), Marsh Creek (n=6) and Walnut Creek (n=3).

3.3 Distribution of Trash Transported

A comparison of the relative proportion of types of trash collected by the three sampling methods was made for Marsh Creek because this is the only study site that was sampled using all three methods. The percentage breakdown for the 11 most common trash types is provided below in Figure 27. From this comparison we can see that the trash trap captures a greater fraction of styrofoam and cigarette butts, but is not effective at capturing film, food wrappers and plastic bags that were commonly collected between storms from the grid area. Similarly, only a few food wrappers and no film or plastic bags were observed during the visual counts. We suspect that the bags and film are primarily moving in the subsurface channel flow rather than floating on the surface. Bags and larger pieces of film were frequently found caught on tree limbs, wood and other debris in the floodplain and along the channel bed and banks. The grid samples also include more trash from other categories that were infrequently or never observed floating past such as straws, food containers, glass bottles and personal care products. The visual observation reported a high fraction of hard plastic pieces. It is possible that these small pieces are easily transported so they rarely settled out in the sampling grid and passed through the trash trap.

The total trash counts normalized by sampling effort (Figure 28) reveal that there is probably a large amount of trash that is bypassing the trash trap, especially foam pieces, plastic bottles, hard plastic pieces, food wrappers, cups and balls and toys. This also corroborates the finding that the trash capture did not increase with higher flows for Marsh Creek. This is likely because trash is bypassing the trap when large wood or debris is captured and blocks the basket. This conclusion is further confirmed by video we collected of styrofoam and plastic bottles jumping over the

boom during high flows at Marsh Creek. In addition, it appears that only a small fraction of the total trash load is being deposited on the floodplain or channel bed or entangled in debris in the riparian corridor. Rather, the bulk of trash washing into our streams is plastics and is being transported rapidly downstream during high flows and is likely to continue being transported to the mainstem of the Neuse River where it will continue to the Pamlico Sound. We can also conclude that stream-clean ups and trash collection, the most common ways to estimate and manage plastics in waterways, severely underestimate the number of beverage bottles, plastic pieces, plastic cups and balls/toys that are transported in urban waterways. *Therefore, programs to prevent this litter from being deposited on the ground and washed into the stormdrain system are critical to plastics from entering stormwater systems in urban areas and being transported to downstream rivers and estuaries of critical social, economic and environmental importance.*



Figure 27 Distribution of trash collected from Marsh Creek by traps (n=15), Grid (n=5) and Visual (n=6) sampling methods.



Figure 28. Number of pieces of litter normalized by the sampling effort for trash traps (n=15), Grid (n=5) and Visual (n=6).

3.4 Outreach

Development and implementation of this project involved collaboration among a number of project partners including NC Sea Grant, NC State University's Biological & Agricultural Engineering Department, the Plastics Ocean Project, Sound Rivers, Inc. and UNC-Wilmington's Department of Chemistry & Biochemistry. This group worked together to design the study, conduct the sampling, troubleshoot issues with the laboratory analyses, and interpret the data results. In addition to the sampling that was outlined and funded by this project, we also participated in the 100 Plastic Rivers project that was led by the University of Birmingham in the U.K. Both water and sediment samples were collected from all 15 of our study sites and sent them to the U.K. for plastics identification. Water samples were collected using a bailing method that required filtering 100 liters of water through a 64 µm sieve. In addition, approximately 100 grams of sediment were collected along the edge of the stream just below the water line. The results from this collaboration allowed us to make broader conclusions about the data from our microplastics sampling conducted with the larger mesh (335 μ m) neuston net. These results indicated that less than 10% of the microplastic particles captured using the 64 µm were larger than 335 µm mesh in our study streams. This collaboration revealed that our trawl net sampling has likely substantially underestimated the microplastic presence in our waterways.

Our results and methods have been shared in a number of ways to inform other research projects and raise awareness of how the presence of plastics in our rivers. To reach a wide audience of citizens and special interest groups such as anglers, an article about plastics entitled, "Plastic, Plastic, Everywhere" was featured in the winter 2020 issue of the NCSG *Coastwatch* magazine. Details of the study and preliminary results were also presented at two webinars ("Tell Me about It Tuesdays" hosted by Sound Rivers, Inc. (10 participants) and for a Xylem corporation

volunteer program (300+ participants)). To reach other researchers, water resource managers, and policy experts, the results of the project were presented at both the 2021 and 2022 NC Marine Debris Symposium (over 100 total attendees) and during a special session on plastics at the 2022 NC Water Resources Research Institute Annual Conference (approximately 120 attendees). In addition, a draft manuscript has been developed featuring the results of the microplastics work, in collaboration with the researchers from the U.K. Birmingham. This article has been submitted to an academic journal for consideration for publication. More informally, our results have been conveyed through email with the City of Raleigh stormwater staff.

4 Conclusions, Recommendations and Lessons Learned

This research provides some of the first quantification of the concentration of micro and macroplastics in North Carolina's rivers, and an estimate of the microplastics loading rates to the Pamlico Sound from the Neuse River. Microplastics were sampled at 15 locations across a large river basin to examine spatial, land cover, and streamflow related impacts on microplastic concentrations. Microplastics were found in all water samples, but the concentrations and polymer composition varied widely between samples. Median microplastic concentration was correlated with land cover for the tributary catchments and the highest microplastic concentrations were observed in urban streams during high streamflow.

In the case of trawl sampling, we found that the level of effort required to quantify microplastic concentrations may not be advisable given the probable substantial underestimation of this approach. The large amount of organic matter (leaves, pollen, etc.), and sediment collected with most samples required time consuming and expensive chemical and physical processes to separate the plastics from the natural materials. In addition, it is possible that plastic particles, particularly fibers, may be lost during this process. This conclusion is supported by the result of concentrations measured from bailing samples collected for the 100 Rivers Project that were one to two orders of magnitude higher than the samples we collected using the 335 μ m mesh; the ratio of concentrations of 64 μ m samples to 335 μ m samples ranged from 38 to 333 (r=0.19). In fact, we estimated the load to be about 230 billion particles per year based on the results from the bailing samples (335 μ m). Using a finer mesh size and sampling a smaller volume, allows for less intensive processing and may provide a better estimate of the number of microplastics. However, with this strategy, the volume of water collected will be substantially reduced, which may risk not capturing all the types of microplastic particles present (Tamminga et al., 2022).

Our work to sample macroplastics indicates that to capture the range of trash types transported through stream networks, it is necessary to employ multiple sampling techniques. Grid sampling will underestimate the proportion of floatable trash (styrofoam, plastic bottles, plastic pieces, etc.). Trapping devices and visual counts during storm flow will underestimate bags, film, wrappers and other trash that tends to move lower in the water column rather than closer to or floating on the surface. In addition, the catchment devices only catch a portion of the trash and are less successful at trash capture during high flow due to clogging with large woody debris, and from bypass during turbulent flows.

We echo the sentiments of other researchers (e.g., Kapp & Yeatman, 2018; Lenaker et al., 2019) in calling for the development of standard procedures for microplastic collection, processing and analysis. The variability in sampling and analysis methods limits the comparability of results and contributes to uncertainty in quantifying the presence of microplastics in the aquatic environment.

Given our observations of increasing microplastic concentration and macroplastic volumes associated with high stream discharge in urban streams moving during higher flows, we suspect that both macroplastics and microplastics in urban areas wash off and move downstream as pulses. More work is needed to study how both micro- and macroplastics concentrations and volumes change during run off events and how the mixing (or lack thereof) occurs when runoff from urban areas enters larger downstream tributaries.

This research project conducted the most in-depth examination of plastics in a North Carolina riverine system and provided not only a better understanding of the presence of this type of pollution but equipped us with a deeper understanding of the effectiveness of sampling methods. Our acquired knowledge will help to inform both local and state level programs in selecting strategies to monitor plastic pollution as well as bolster efforts across the coast to quantify the scale of the problem and take meaningful action.

5 Acknowledgements

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6 Data Availability

The data generated by this project has been submitted to the NOAA National Centers for Environmental Information (NCEI). Metadata with instructions for data use and interpretation have also been provided. The collected data and sample collection methodologies are also available to state, federal, local, and tribal entities, as well as to the general public, upon request by contacting Dr. Barbara Doll at bdoll@ncsu.edu. This report has been submitted to the NOAA Institutional Repository at http://library.noaa.gov/repository.

7 References and Literature Cited

- Abreu, A., & Pedrotti, M. L. (2019). Microplastics in the oceans: the solutions lie on land. *Field Actions Science Reports. The journal of field actions*(Special Issue 19), 62-67.
- Arthur, C., Baker, J. E., & Bamford, H. A. (2009). Proceedings of the International Research Workshop on the Occurrence, Effects, and Fate of Microplastic Marine Debris, September 9-11, 2008, University of Washington Tacoma, Tacoma, WA, USA.
- Baldwin, A. K., Corsi, S. R., & Mason, S. A. (2016). Plastic debris in 29 Great Lakes tributaries: relations to watershed attributes and hydrology. *Environmental science & technology*, 50(19), 10377-10385.
- Choy, C. A., Robison, B. H., Gagne, T. O., Erwin, B., Firl, E., Halden, R. U., Hamilton, J. A., Katija, K., Lisin, S. E., & Rolsky, C. (2019). The vertical distribution and biological transport of marine microplastics across the epipelagic and mesopelagic water column. *Scientific reports*, 9(1), 7843.
- De Sá, L. C., Oliveira, M., Ribeiro, F., Rocha, T. L., & Futter, M. N. (2018). Studies of the effects of microplastics on aquatic organisms: what do we know and where should we focus our efforts in the future? *Science of the total environment*, *645*, 1029-1039.
- De Stephanis, R., Giménez, J., Carpinelli, E., Gutierrez-Exposito, C., & Cañadas, A. (2013). As main meal for sperm whales: plastics debris. *Marine pollution bulletin*, *69*(1-2), 206-214.
- Doll, B. A., Wise-Frederick, D. E., Buckner, C. M., Wilkerson, S. D., Harman, W. A., Smith, R. E., & Spooner, J. (2002). Hydraulic geometry relationships for urban streams throughout the piedmont of north carolina. *Journal of the American Water Resources Association*, 38(3), 641-651.
- Dris, R., Gasperi, J., Rocher, V., Saad, M., Renault, N., & Tassin, B. (2015). Microplastic contamination in an urban area: a case study in Greater Paris. *Environmental Chemistry*, 12(5), 592-599.
- Geyer, R., Jambeck, J. R., & Law, K. L. (2017). Production, use, and fate of all plastics ever made. *Science advances*, *3*(7), e1700782.
- González-Fernández, D., & Hanke, G. (2017). Toward a harmonized approach for monitoring of riverine floating macro litter inputs to the marine environment. *Frontiers in Marine Science*, *4*, 86.
- Gray, A. D., Wertz, H., Leads, R. R., & Weinstein, J. E. (2018). Microplastic in two South Carolina Estuaries: Occurrence, distribution, and composition. *Marine pollution bulletin*, *128*, 223-233.
- Jambeck, J. R., Geyer, R., Wilcox, C., Siegler, T. R., Perryman, M., Andrady, A., Narayan, R., & Law, K. L. (2015). Plastic waste inputs from land into the ocean. *Science*, 347(6223), 768-771.
- Kapp, K. J., & Yeatman, E. (2018). Microplastic hotspots in the Snake and Lower Columbia rivers: A journey from the Greater Yellowstone Ecosystem to the Pacific Ocean. *Environmental pollution*, 241, 1082-1090.
- Kershaw, P. J. (2016). Marine plastic debris and microplastics–Global lessons and research to inspire action and guide policy change.
- Koelmans, A. A., Besseling, E., & Foekema, E. M. (2014). Leaching of plastic additives to marine organisms. *Environmental pollution*, 187, 49-54.
- Lenaker, P. L., Baldwin, A. K., Corsi, S. R., Mason, S. A., Reneau, P. C., & Scott, J. W. (2019). Vertical distribution of microplastics in the water column and surficial sediment from the

Milwaukee River Basin to Lake Michigan. *Environmental science & technology*, 53(21), 12227-12237.

- Lin, L., Zuo, L.-Z., Peng, J.-P., Cai, L.-Q., Fok, L., Yan, Y., Li, H.-X., & Xu, X.-R. (2018). Occurrence and distribution of microplastics in an urban river: a case study in the Pearl River along Guangzhou City, China. *Science of the total environment*, 644, 375-381.
- Mascarenhas, R., Santos, R., & Zeppelini, D. (2004). Plastic debris ingestion by sea turtle in Paraíba, Brazil. *Marine pollution bulletin*, 49(4), 354-355.
- Mendoza, L. M. R., & Balcer, M. (2019). Microplastics in freshwater environments: a review of quantification assessment. *TrAC Trends in Analytical Chemistry*, *113*, 402-408.
- Moore, C. J., Lattin, G. L., & Zellers, A. (2011). Quantity and type of plastic debris flowing from two urban rivers to coastal waters and beaches of Southern California. *Revista de Gestão Costeira Integrada-Journal of Integrated Coastal Zone Management*, 11(1), 65-73.
- MRLC. (2022). *The Multi-Resolution Land Characteristics (MRLC) Consortium*. https://www.mrlc.gov/
- Munier, B., & Bendell, L. (2018). Macro and micro plastics sorb and desorb metals and act as a point source of trace metals to coastal ecosystems. *PLoS One*, *13*(2), e0191759.
- Rochman, C. M., Hoh, E., Hentschel, B. T., & Kaye, S. (2013). Long-term field measurement of sorption of organic contaminants to five types of plastic pellets: implications for plastic marine debris. *Environmental science & technology*, 47(3), 1646-1654.
- Teuten, E. L., Saquing, J. M., Knappe, D. R., Barlaz, M. A., Jonsson, S., Björn, A., Rowland, S. J., Thompson, R. C., Galloway, T. S., & Yamashita, R. (2009). Transport and release of chemicals from plastics to the environment and to wildlife. *Philosophical transactions of the royal society B: biological sciences*, 364(1526), 2027-2045.
- Thompson, R. C. (2015). Microplastics in the marine environment: sources, consequences and solutions. In *Marine anthropogenic litter* (pp. 185-200). Springer, Cham.
- United States Census Bureau Data (2022). https://www.census.gov/data.html
- Van Emmerik, T., Kieu-Le, T.-C., Loozen, M., Van Oeveren, K., Strady, E., Bui, X.-T., Egger, M., Gasperi, J., Lebreton, L., & Nguyen, P.-D. (2018). A methodology to characterize riverine macroplastic emission into the ocean. *Frontiers in Marine Science*, 5, 372.
- Vorkinn, M., & Riese, H. (2001). Environmental concern in a local context: The significance of place attachment. *Environment and Behavior*, *33*(2), 249-263.
- Yonkos, L. T., Friedel, E. A., Perez-Reyes, A. C., Ghosal, S., & Arthur, C. D. (2014). Microplastics in four estuarine rivers in the Chesapeake Bay, USA. *Environmental science & technology*, 48(24), 14195-14202.
- Zettler, E. R., Mincer, T. J., & Amaral-Zettler, L. A. (2013). Life in the "plastisphere": microbial communities on plastic marine debris. *Environmental science & technology*, 47(13), 7137-7146.

8 Appendix

8.1 Litter Collection Data Sheet

| | Organization | Name of organization responsible for data collection |
|--|---------------|--|
| STREAM DEBRIS Debris Density Data Sheet | Surveyor name | Name of person responsible for filling in this sheet |
| | Phone number | Phone contact for surveyor |
| Complete this form during | Email address | Email contact for surveyor |
| EACH survey per site visit | Survey Date | Date of this survey |

ADDITIONAL INFORMATION

| Stream name | | | |
|--------------------------------|--------------------|-----|---|
| Survey Type | Accumulation | | |
| Time start/end | Start | End | Time at the beginning and end of the survey |
| Number of persons | | | Number of persons conducting the survey |
| Photos | | | Did you take photos of any debris items during this survey? |
| Notes: Evidence of cleanup, st | urvey issues, etc. | | |

| Site Name | | | | | | | PAGE 2 of 3 | | |
|-------------|--|----------------|------|----|----------------------|--|-------------|--|--|
| Survey Date | | | | | | | | | |
| | ITEM | TALLY (e.g., T | | | | | | | |
| 1 | Plastic pieces / fragments | | Hard | PL | ASTIC Foam | | Film | | |
| 2 | Beverage Bottles | | | | | | | | |
| 3 | Food Wrappers & Labels (chip /candy, etc.) | | | | | | | | |
| 4 | Bags (whole or most) | | | | | | | | |
| 5 | Auto Fluid Containers (oil, antifreeze, etc) | | | | | | | | |
| 6 | Other Jugs or Containers (cleaning, buckets) | | | | | | | | |
| 7 | Cups (plastic & foam) | | | | | | | | |
| 8 | Lids & Bottle Caps | | | | | | | | |
| 9 | Cigar tips, Cigarette Butts | | | | | | | | |
| 10 | Disposable Lighters | | | | | | | | |
| 11 | Rope/ Net Pieces | | | | | | | | |
| 12 | Fishing Lures & Line | | | | | | | | |
| 13 | Food Containers (clam shells) Foam or Plastic | | | | | | | | |
| 14 | Utensils | | | | | | | | |
| 15 | Straws | | | | | | | | |
| 16 | Balloons Toys or Sports | | | | | | | | |
| 17 | Equipment (e.g., balls) | | | | | | | | |
| 18 | Personal Care Products (floss, brushes, etc) | | | | | | | | |
| 19 | Automotive Parts | | | | | | | | |
| 20 | Pipes / Conduit | | | | | | | | |
| 21 | Safety or Survey | | | | | | | | |
| 22 | Shipping (bubble wrap) | | | | | | | | |
| 23 | Other Plastic: | | | | | | | | |

| Site Name | | | PAGE 3 of 3 | | | | |
|------------------------------------|----------------------------|----------------------|-------------|----------|--|--|--|
| Survey Date | | | | | | | |
| ITEM | TALLY (e.g., M) | | | | | | |
| | | | | | | | |
| | RUBBE | CR | | | | | |
| 24 Gloves | | | | | | | |
| 25 Shoes (flip flop, etc) | | | | | | | |
| 26 Tires | | | | | | | |
| 27 Rubber Strips | | | | | | | |
| 28 Rubber fragments | | | | | | | |
| 29 Other: | | | | | | | |
| | META | L | | | | | |
| | | | | | | | |
| 30 Aluminum Can | | | | | | | |
| | | | | | | | |
| 31 Tin Can | | | | | | | |
| 32 Aerosol cans | | | | | | | |
| 33 Metal fragments | | | | | | | |
| 34 Other: | | | | | | | |
| | GLAS | S | | | | | |
| 35 Beverage bottles | | | | | | | |
| 36 Jars | | | | | | | |
| | | | | | | | |
| 37 Glass fragments | | | | | | | |
| | | | | | | | |
| 38 Other: | | | | | | | |
| Steller. | CLOTH/FA | RRIC | | | | | |
| 39 Clothing towels fabric | CLOIII/IA | DAIC | | 1 | | | |
| ss clothing, towers, faorie | DADE | P | | | | | |
| 40 Cardboard/naper | 1711 | Α | | | | | |
| hags/naper | | | | | | | |
| loags/paper | OTHER UNCLASSIFIE | D TRASH / NOTES | | | | | |
| 41 | OTHER OTCLASSIFIEL | 7 1121511 / 1101L5 | | 1 | | | |
| | | | | | | | |
| | | | | | | | |
| LARGE DEB | RIS ITEMS > 2 feet, not in | another category and | REMOVED | | | | |
| Describe Item (vessel, chair etc.) | Approximate size | Location | Photo ID | , if any | | | |
| 42 | | | | | | | |
| 42 | | | | | | | |
| | | | | | | | |
| | | | OT DEMOVED | | | | |
| LARGE DEBRI | STIEMS > 2 feet, not in an | other category and N | OT REMOVED | | | | |
| Describe Item (vessel, chair etc.) | Approximate size | Location | Photo ID | , if any | | | |
| | | | | | | | |
| 43 | | | | | | | |
| | | | | | | | |
| | | | | | | | |
| | | | | | | | |

| Plastic Bottles | Weight (grams) |
|------------------------------|----------------|
| 1.7 oz Mini liguor | 12 |
| 1.93 oz 5 hour energy | 12.5 |
| 8 oz water | 8.5 |
| 8 oz soda | 11.5 |
| 8 oz Mini-water bottle | 19 |
| 8 oz Juice | 27 |
| 10 oz Juice | 16 |
| 10 oz Gatorade | 17.5 |
| 16 oz Snapple with lid | 55 |
| 10 oz Soda (Ginger ale) | 16 |
| 12 oz Soda | 13 |
| 12 oz Minute Maid | 33.5 |
| 12 oz Gatorade | 22 |
| 12 oz Liquor | 23 |
| 15.2 oz Minute Maid | 39.5 |
| 16 oz Water (thin plastic) | 10 |
| 16 oz Soda | 26 |
| 16 oz Water with pull up top | 37.5 |
| 16 oz Gatorade | 32.5 |
| 20 oz Water | 26 |
| 20 oz Soda | 26 |
| 20 oz Gatorade | 35 |
| 22 oz Propel Bottle | 26 |
| 24 oz Soda | 36.5 |
| 24 oz Water | 36.5 |
| 28 oz Powerade | 44.5 |
| 28 oz Gatorade | 44 |
| 28 oz Soda | 28 |
| 28 oz Water | 28 |
| 30.4 oz Core Hydration | 69 |
| 32 oz Gatorade | 52 |
| 32 oz Water | 32 |
| 34 oz Water | 34 |
| 34 oz Soda | 34 |
| 36 oz Soda | 36 |
| 40 oz Plastic Beer | 70 |
| 42 oz Ice house w/cap | 70 |
| 1 Liter soda w/cap | 39 |
| 1 Liter soda w/out cap | 36 |
| 1 Liter Juice | 48 |
| 2 Liter Soda w/cap | 55 |
| 1 Gallon Milk Jug | 62 |

8.2 Library of Plastic Bottle Weights